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GAMMA-GAMMA DIRECTIONAL CORRELATIONS IN THE DECAY OF Ir^{192}

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GAMMA-GAMMA DIRECTIONAL CORRELATIONS IN THE DECAY OF Ir^{192}

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SUMMARY

The decay of Ir^{192} yields a number of composite gamma peaks, each of which contains several gamma rays very closely spaced in energy. These are the 600-, the 470-, the 300-, and the 200-keV composite gamma peaks. In the previous investigations, most of the gamma-gamma directional correlation measurements have been made employing thalium activated sodium iodide gamma detectors [NaI(Tl)]. The energy resolution of these detectors is poor; therefore, these detectors cannot be used to select a single gamma ray out of a group of several radiations of about the same energy. Also, gamma rays of low intensity which have energies fairly close to the energies of intense radiations could not be selected. Thus some previous gamma-gamma directional correlation measurements were made on composite cascades of gamma rays. The contribution of each of the constituents of the cascades was then estimated by using the measured values of the gamma intensities and energy resolution of the NaI(Tl) detectors.

This research on gamma-gamma directional correlation measurements in the decay of Ir^{192} is undertaken employing a liquid nitrogen cooled lithium drifted germanium gamma detector of 30 cm^3 volume for one radiation and a conventional 3 X 3 inch NaI(Tl) detector for the other radiation. The use of a Ge(Li) detector permits gamma-gamma directional correlation experiments to be done directly on the individual gamma rays that are a part of the composite peaks.

In the present work three gamma detectors are used: a fixed Ge(Li) detector and two movable 3 X 3 inch NaI(Tl) crystals, D_1 and D_2. In some experiments, lead is placed on the faces of D_1 and D_2 to reduce the possibility of coincident summing. A standard fast-slow coincidence circuit is employed with a resolving time of about 20 nanoseconds. A single-channel analyzer is used in each of the NaI(Tl) detectors and a 400-channel analyzer is used in two input operation on the Ge(Li) detector. Two triple, slow coincidence circuits produce two signals from two fast coincidence circuits and two single-channel analyzers. These slow outputs provide gating signals for each of the input sections of the 400-channel analyzer. The 400-channel analyzer then displays the two expanded coincidence spectra. Thus two essentially independent experiments were done simultaneously.

The Ir^{192} was produced by irradiation of iridium with thermal neutrons at the Oak Ridge National Laboratory and furnished in the form of Na_2IrCl_6 in HCl solution. The source was prepared by evaporation to dryness in a plastic container. The gamma rays studied in this research showed no change in relative intensities over a period of about a year; therefore, radioactive impurities are probably not present in appreciable quantities in the source.

In order to determine which gamma-gamma directional correlations were feasible, considerable coincidence work was performed. From these studies it was decided to do the following correlation experiments [gamma energies given in keV]: (468-417), (612-588), (588-296), (612-308), (316-604), (484-206), and (206-374). Measurements were made at angles of 90°, 135°, 180°, 225°, and 270°.

The experimental results were interpreted in terms of the spins of the relevant excited levels of Pt^{192} and Os^{192} and multipolarity assignments of the gamma transitions. The (468-417) keV correlation experiment indicates a spin of 4 for the 1200 keV level in Pt^{192} and a nearly pure E_2 radiation for the 417 keV gamma. Spin 4 for the 1200 keV level is found to be consistent with the results of the (588-612) keV correlation experiment. The assignments of spin 3 to the 690 keV level in Os^{192} and the 921 keV level in Pt^{192} are in agreement with the experimental data on the (484-206), (308-612), and (316-604) keV correlations. The results of the (374-206) keV correlation experiment are consistent with spin 4 for the 580 keV level in Os^{192}. The multipolarities of the 296-, 308-, and 484-keV transitions were determined as predominantly E_2 with an admixture of one or two percent of M_1 radiation. The multipolarity of the 604 keV transition was found to be about 19 percent M_1 plus 81 percent E_2 radiation.

CHAPTER I

INTRODUCTION

At present, there does not exist a well-established theory of the nucleus through which all its properties can be understood. There exist a number of nuclear models or rudimentary theories of restricted validity and the study of the nucleus is largely the study of these models. The well-known models are the shell model (1) and the collective model (2, Chapter X). These models predict the nuclear state properties like energy, spin and parity, and multipolarities of the gamma transitions connecting the various states. The task of the nuclear experimentalist is twofold; first, he checks the predictions of these nuclear models and second, he provides experimental data for reference in the evolution of a satisfactory comprehensive theory of the nucleus.

One method of securing useful experimental data is through study of gamma rays in the decay of radioactive isotopes. This research concerns itself with the experimental study of gamma radiation emitted in the decay of 74 day Ir^{192} through gamma-gamma directional correlation techniques. Preliminary results of this research have been presented at the 1968 meeting of the Southeastern Section of the American Physical Society (3).

The Role of Gamma-Gamma Directional Correlations (2)

The phenomenon of angular correlations involves the cascade emission of two (or more) radiations. The coincidence counting rate is considered as a function of the angle θ between the propagation vectors of the two radiations. Ordinarily, the radiation pattern from a radioactive isotope is isotropic due to the random orientation of the nuclei in space. To observe anisotropy in the radiation pattern of a source, the following procedure is adopted. Let the nuclei decay through two radiations R_1 and R_2 in rapid succession. If R_1 is observed in a fixed direction $\vec{k_1}$, then an ensemble of nuclei having an anisotropic distribution of spin orientations is selected. The radiation R_2 may show a definite angular correlation with respect to $\vec{k_1}$. This means the probability, that two successive radiations will be emitted, may depend on the angle θ between the directions of their emission.

The correlation function $W(\theta)$ is defined as follows:

$W(\theta)\, d\Omega$ is the relative probability that radiation R_2 is emitted into the solid angle element $d\Omega$ at an angle θ measured with respect to $\vec{k_1}$, the propagation direction of radiation R_1.

Hamilton (4) was the first to publish the theory of angular correlations. Early attempts to experimentally verify the theory failed due to inadequate experimental techniques. In 1947, Brady and Deutsch (5) performed the first successful gamma-gamma directional correlation experiments using Geiger counters as radiation detectors. In 1948, Brady and Deutsch (6) used scintillation detectors for directional correlation work, thus considerably improving the counting efficiency, counting speed, and energy resolution. Since 1948, the gamma-gamma directional correlation measurements have become an important tool for investigating the properties of the nucleus. In 1953, Biedenharn and Rose (7) worked out the complete theory of the angular correlations of the nuclear radiations. The correlation function $W(\theta)$ is usually represented as a series of even order Legendre polynomials

$$W(\theta) = 1 + A_2 P_2 (\cos\theta) + A_4 P_4 (\cos\theta) + --- . \qquad (1)$$

The correlation coefficients A_2, A_4 --- have been tabulated by Biedenharn and Rose (7) and Ferentz and Rosenzweig (2) as functions of (i) spins of the nuclear levels, connected by the gamma transitions, R_1 and R_2, and (ii) multipole character of the gamma transitions [Appendix A].

In the performance of a directional correlation experiment, the coincidence counting rates between radiations R_1 and R_2 are measured as functions of the angle θ between the directions of emission. Then the various corrections for the accidental coincidences, Compton background, and finite solid angles subtended by the counters are applied. Finally, the experimentally determined correlation coefficients are compared with the theoretical values to obtain (i) spin values for the relevant nuclear levels and (ii) multipolarities of the gamma transitions.

The Ir^{192} Problem

Gamma-gamma directional correlation measurements in the decay of Ir^{192} have been made by many investigators. Pringle, et al. (8) employed one inch cylindrical NaI(Tl) crystals and studied a few cascades. Baggerly et al. (9) were the first to study the decay of Ir^{192} extensively. They determined the energies, intensities, internal conversion coefficients, and multipolarities of the gamma transitions and investigated the gamma-gamma directional correlations of a few cascades. The spins and parities of most of the levels were given. Kelly, et al. (10) made gamma-gamma directional correlation measurements on a number of isotopes including Pt^{192}. Their assignments of spin values to various excited states and multipole characters to some of the transitions were in rough agreement with those of the previous investigators. Shiel, et al. (11) measured the directional correlations on a number of cascades to remove uncertainties in the values of spins assigned to some of the excited levels in Pt^{192} and Os^{192} and to add evidence in support of the decay scheme of Ir^{192}.

J. Mraz (12) measured gamma-gamma directional correlations for 300 keV cascade of composite gamma rays to remove ambiguities in the spins assigned to the fourth excited level of Pt^{192} and determine the multipole character of the 296- and 308-keV gamma transitions. The investigation of Kawamura, et al. (13) was intended to remove uncertainties in the spin values assigned to the third and fourth excited states of Pt^{192} by further measurements of gamma-gamma directional correlations, corrected more precisely for the interference effects caused by the high energy gamma rays. In their intensive gamma-gamma directional correlation work, Simons, et al. (14) included the study of some weak transitions. A number of possible spin values for the highest level, 1380 keV, were suggested. The spin values of 3^+ and 4^+ were assigned to 921- and 1200-keV excited levels of Pt^{192}, respectively. This assignment was subsequently supported by the gamma-gamma directional correlation measurements of Johns, et al. (15). Johns, et al. also determined the multipolarities of the higher energy gamma transitions in Pt^{192}.

Koch, et al. (16) measured the directional correlations of the (2-2-0) cascades in a number of nuclei including Pt^{192} and determined the M_1/E_2 mixing ratio for the 296 keV gamma transition. Kumar (17) used sum and sum peak coincidence scintillation spectrometers for gamma-gamma directional correlation work to check on the spin assignments 4^+ for the 1200 keV excited level in Pt^{192}, among other things.

Apart from the gamma-gamma directional correlation work, a number of other investigations were carried out on the decay of Ir^{192}. Johns, et al. (18) measured the energies and intensities of most of the gamma rays in the decay of Ir^{192} by studying the external conversion spectra with a high-resolution beta-ray spectrometer. A prism beta-spectrometer was employed by Kelman, et al. (19) to study the conversion electron spectrum of Pt^{192} and Os^{192}. All investigated transitions were found to be of E_2 or $E_2 + M_1$ character and the M_1 admixtures were determined. V. Shiel (20) did some beta-gamma correlation work on Ir^{192} and certain facts supporting the decay scheme were verified.

Electron-gamma correlation measurements were made by Butt, et al. (21) to determine the mixtures of 296- and 308-keV transitions of Pt^{192}. The relative gamma intensities and energies in Ir^{192} were measured by Lindstrom, et al. (22) with a bent-crystal spectrometer and a double focussing beta spectrometer. The decay scheme of Ir^{192} was discussed. Using a double focussing beta spectrometer, Schellenberg, et al. (23) studied internal and external conversion spectra in the decay of Ir^{192}. New multipolarity information was obtained for the higher energy gamma transitions and modifications of the previously reported decay schemes for Pt^{192} and Os^{192} were proposed. In the following year, Schellenberg, et al. (24) and Palaska, et al. (25) employed nitrogen cooled lithium drifted germanium gamma detectors and measured the gamma ray intensities and energies in the decay of Ir^{192}. Minor modifications in the decay schemes were proposed.

Beta-gamma correlation measurements on Ir^{192} were made by Bhattacherjee, et al. (26). In this publication the authors deduced values of the mixing ratios in the 296-, 308-, and 604-keV gamma transitions which gave reasonably good fits to the results of their experiments. To reduce certain involved complexities in the determination of the mixing ratio for the 604 keV gamma transition, they suggested gamma-gamma directional correlation work on the (300-600) keV cascades with one or more cascades eliminated.

Purpose of This Research

Previously, most of the gamma-gamma directional correlation measurements in Ir^{192} were made employing thalium activated sodium iodide gamma detectors [NaI(Tl)]. The energy resolution of these detectors is poor; therefore, these detectors could not be used to select a single gamma ray out of a group of several radiations of about the same energy. Also, gamma rays of low intensity which have energies fairly close to the energies of intense radiations could not be selected. The decay of Ir^{192} [Figure 1] yields a number of composite gamma peaks, each of which contains several gamma rays very closely spaced in energy, namely, the 600-, the 470-, the 300-, and the 200-keV composite gamma peaks. Thus some previous gamma-gamma directional correlation measurements in Ir^{192} were made on composite cascades of gamma rays. The contribution of each

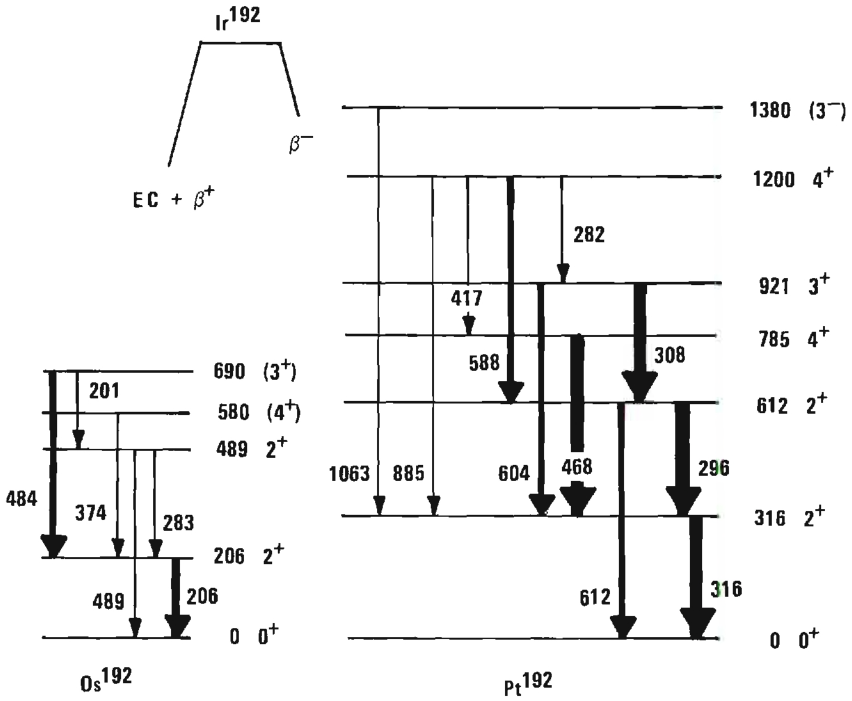

Figure 1. Decay Scheme of Ir^{192} According to Palaska, et al. (25)

of the constituent cascades was estimated by using the measured values of the gamma intensities and from a knowledge of the energy resolution of the NaI(Tl) detectors.

This research on gamma-gamma directional correlation measurements in Ir^{192} is undertaken employing a liquid nitrogen cooled lithium drifted germanium gamma detector of 30 cm^3 volume for one radiation and a conventional 3 x 3 inch NaI(Tl) detector for the other radiation. The use of a GeLi detector permits gamma-gamma directional correlation experiments to be made directly on individual gamma rays that are closely spaced in energy without many of the uncertainties associated with the use of two NaI(Tl) detectors.

The spins of levels at 1200-, 580-, and 690-keV and multipolarities of 296-, 308-, 374-, 417-, 484-, and 604-keV gamma rays were in some doubt. The principal purposes of this research are:

(i) to check on the values of the spins assigned to certain excited levels and remove uncertainties where possible

(ii) to fix limits on the multipolarity mixing ratios of certain gamma transitions.

CHAPTER II

EXPERIMENTAL APPARATUS

The experimental apparatus used in the present research consisted of scintillation and solid state detectors, amplifiers, pulse height analyzers, coincidence circuits, and data recording equipment (27). A block diagram of the experimental apparatus is shown in Figure 2. In the electronic circuitry, the fast-slow coincidence circuits were employed to study the coincidences between radiations of selected energies. Two essentially independent experiments were done simultaneously.

Gamma Detectors

A 30 cm^3 liquid nitrogen cooled lithium drifted germanium gamma detector [Ge(Li)] is employed to detect one of the radiations in a gamma-gamma cascade. This detector, model LGC.5X, manufactured by Nuclear Diodes Incorporated, is operated at about 1250 volts. The shape of the Ge(Li) is trapezoidal with an active area of 10 cm^2, length 26.5 mm, and drifted depth 11 mm. The energy resolution* of the Ge(Li) is about 1.5 percent for the 316 keV gamma in the decay of Ir^{192}.

Two 3 x 3 inch NaI(Tl) gamma detectors, manufactured by Nuclear Diodes Incorporated, were employed for the second radiation in the cascade. These detectors were called D_1 and D_2. The NaI(Tl) crystals were

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*Resolution is defined as the full width in keV, at half maximum divided by the energy (keV) of the gamma peak.

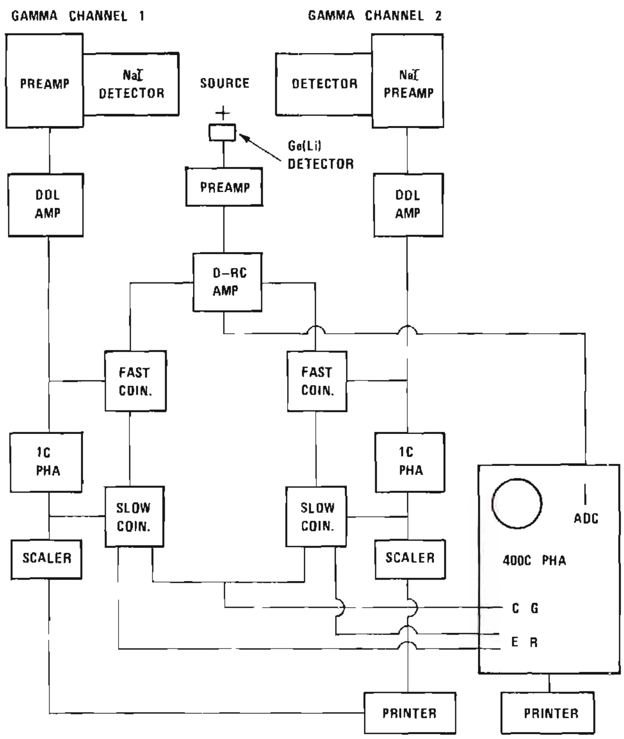

Figure 2. Block Diagram of the Electronic Instrumentation

optically coupled to RCA Type 8054 photomultiplier tubes. These NaI(Tl) detectors were provided with mu-metal magnetic shields and were preassembled by the manufacturer. The operating voltage for the photomultiplier tubes was about 1075 volts. One of these photomultiplier tubes [D_2] required extra magnetic shielding [a few conetic and netic layers].

Counting Geometry

A photograph of the gamma detectors as set up for gamma-gamma directional correlation work is shown in Figure 3. The Ge(Li) was fixed and the two NaI(Tl) detectors were manually rotated in a semicircle about the radioactive source in order to obtain data at different angles θ. The angle θ is the angle between the source-Ge(Li) axis and the axis of one of the NaI(Tl) detectors. In order to minimize any scattering effects, lead shielding of thickness about 8 mm and length about 10 cm was placed concentrically about each NaI(Tl) detector so that "cross-talk" between detectors was much inhibited. No shield was found necessary for the Ge(Li).

Electronics

A block diagram of the electronic instrumentation is shown in Figure 2. A 400-channel pulse height analyzer [Packard Instrument Company Model 116] with coincidence gating was employed as an integral part of the fast-slow coincidence circuitry (28). The 400-channel analyzer is utilized in two input operation on the Ge(Li) with the help of external routing signals. Thus the 400-channel analyzer was used with both detector systems, NaI(Tl)D_1 - Ge(Li) and NaI(Tl)D_2 - Ge(Li), in carrying out two simultaneous correlation experiments. The fast coin-

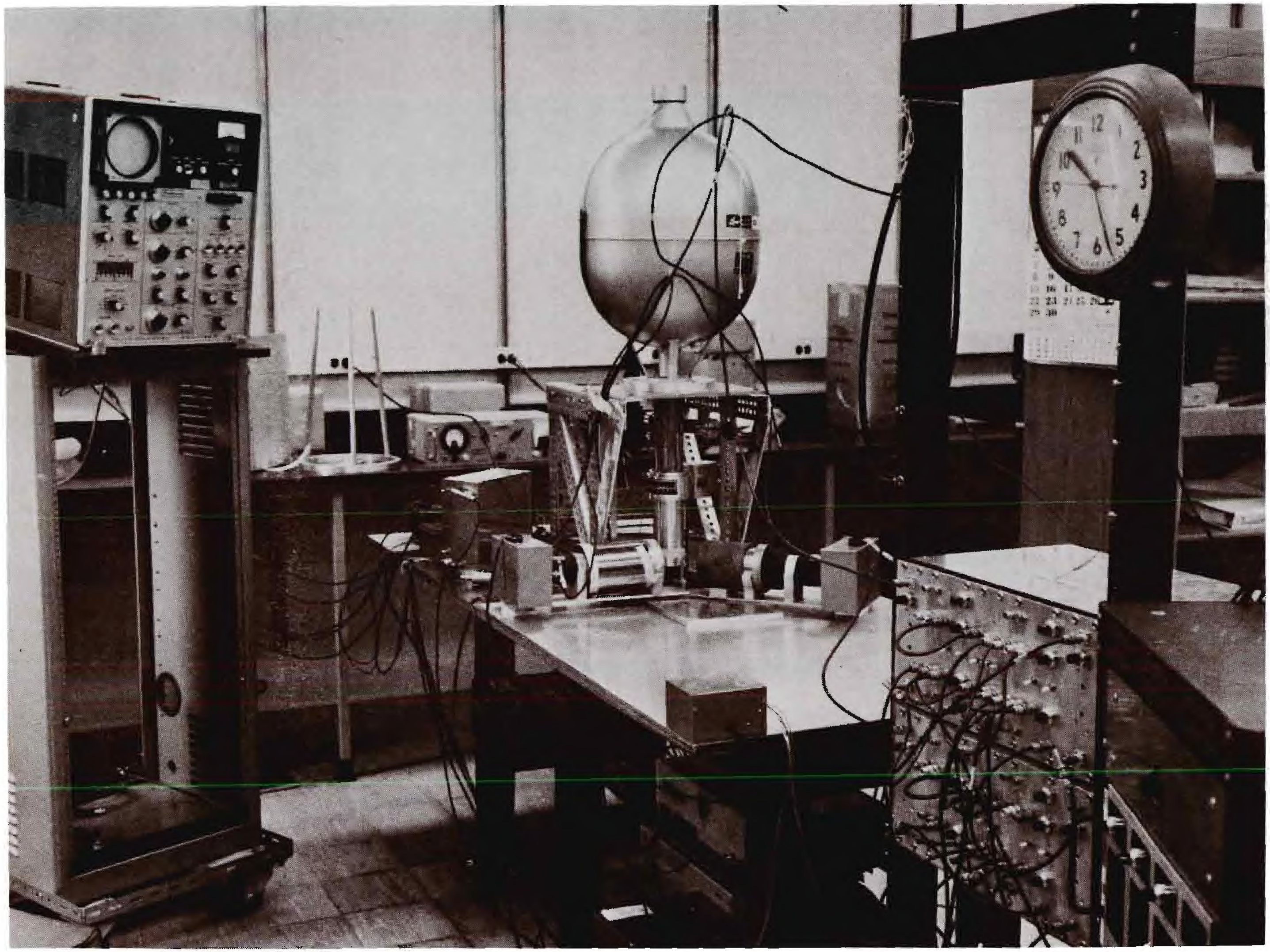

Figure 3. Photograph of the Detectors during a Gamma-Gamma
Directional Correlation Experiment

cidence timing analysis and slow coincidence gamma ray energy selection for the NaI(Tl) detectors were performed in circuits external to the 400-channel analyzer as shown in Figure 3. This part of the instrumentation was designed, constructed, and maintained by Professor E. T. Patronis, Jr.

Double RC clipped and double-delay line main amplifiers provided bi-polar pulses for coincidence timing and energy analysis. Variable electronic delays were inserted in the fast coincidence circuits to compensate for differences in transit times of the signals through the gamma detection system. Both coincidence systems were operated with a resolving time of about 20 nanoseconds. The gamma ray energy analysis for NaI(Tl) detectors was carried out in single-channel pulse height analyzers. Double coincidence signals, carrying both the gamma energy and fast timing information, operated the coincidence gating and external routing inputs of the 400-channel analyzer.

The Ge(Li) signals from a Tennelec preamplifier type TC-135 were further amplified in a double RC clipped main amplifier and then fed to the 400-channel analyzer for pulse height analysis in the two 200-channel memory groups. These signals were gated into the analyzer memory to form a triple coincidence spectrum when they were coincident with a double coincident signal applied at the coincidence gate. These triple coincidences were routed into separate halves of the memory depending on which of the two NaI(Tl) detectors gave a pulse coincident with the pulse from the Ge(Li) detector. The coincidence spectra were accumulated for a preset period of time and then printed directly from the memory on paper tape. The number of counts from the single-channel analyzers used on the NaI(Tl) detectors was recorded hourly.

CHAPTER III

DIRECTIONAL CORRELATION MEASUREMENTS AND RESULTS

Ir^{192} was prepared at the Oak Ridge National Laboratories by irradiation of iridium with thermal neutrons. It was furnished in the form of Na_2IrCl_6 in HCl solution. The source used in the experiment was evaporated to dryness and enclosed in a plastic container of wall thickness about 1 mm and diameter about 10 mm. The gamma rays, studied in this work, showed no change in relative intensities over a period of about a year indicating that radioactive impurities are not present in appreciable quantities in the source. The source strength was about one millicurie.

Singles Spectrum

The prominent feature of the experimental setup of the present research was the high resolution of the Ge(Li) detector. Figure 4a shows the gamma ray singles spectrum of Ir^{192} as viewed by the Ge(Li) detector. The values of the energy resolution at 316 keV and 604 keV were about 1.5 percent and 1.1 percent, respectively. Figure 4b shows the gamma ray singles spectrum as displayed by the NaI(Tl) detector. The singles spectrum was helpful in determining the feasibility of the gamma-gamma directional correlation experiments performed in this research.

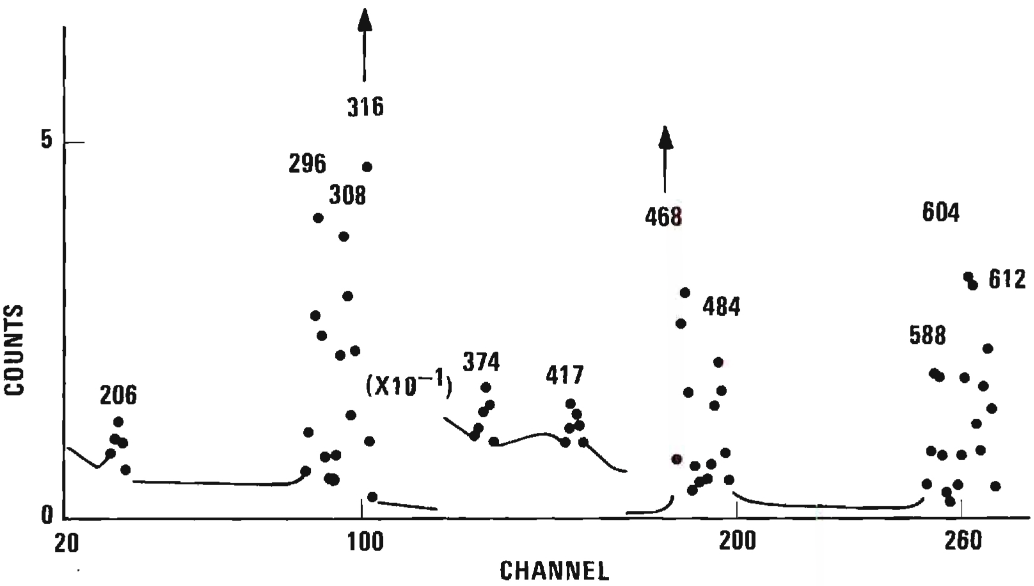

Figure 4a. Gamma Ray Singles Spectrum of Ir^{192} by Ge(Li) Detector

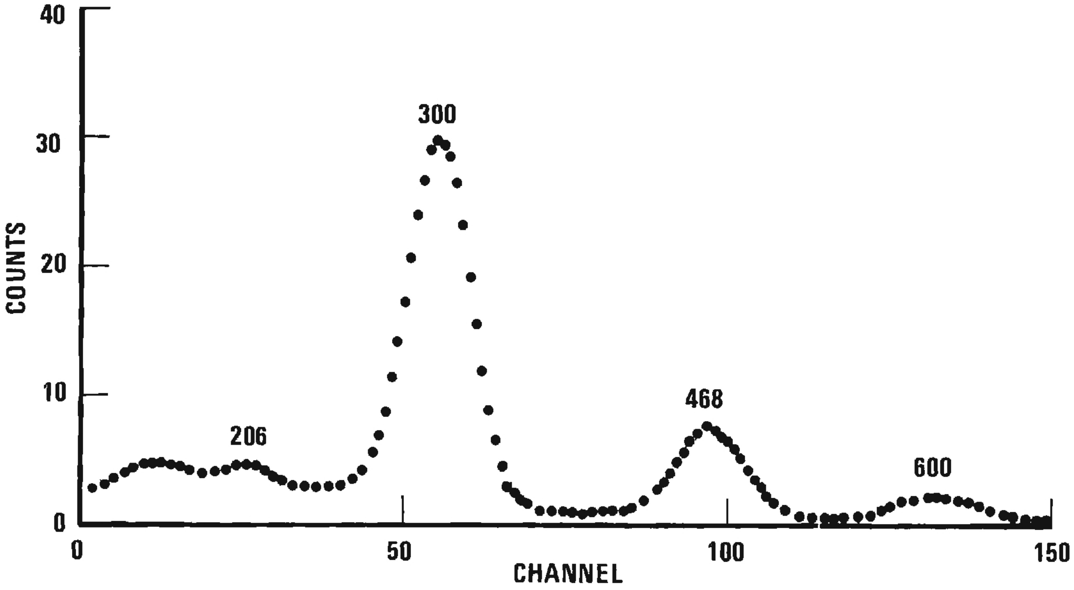

Figure 4b. Gamma Ray Singles Spectrum of Ir^{192}
by NaI(Tl) Detector

Coincidence Spectra

In order to determine which gamma-gamma directional correlations were feasible, it was necessary to perform considerable coincidence work. This type of study helped to find out (i) the coincidence rate and (ii) the effect of other [undesired] cascades on the ones of interest. It sometimes happens that a peak of a weak gamma in a singles spectrum is almost hidden by the background formed because of the Compton scattering in the detector due to higher energy radiations (20,29); 374-, 417- and 206-keV gamma peaks are pertinent examples in the decay of Ir^{192}. However, in some cases, the coincidence spectra [Figure 5a] show up gamma transitions which cannot be easily observed in the singles spectrum.

To observe a coincidence spectrum, an NaI(Tl) detector was set on a resolved gamma peak and the coincidence spectrum in the Ge(Li) detector was displayed on the 400-channel analyzer. In order to obtain coincidence data rapidly, it was necessary to place the gamma detectors close to the source and to use a strong source.

The strength of the source, which could be most efficiently used was determined by the resolving time of the fast-slow coincidence circuit. A resolving time of 20 nanoseconds could be used. With shorter resolving times the real coincidence rate began to decrease thus indicating that all the real coincidences were not being counted. The accidental coincidences are given by the formula: $A = 2\tau N_1 N_2$, where τ is the resolving time and N_1 and N_2 are the singles rates of the two detectors. For a fixed resolving time, it can be shown that an increase in the source strength beyond a point which gives about the same number

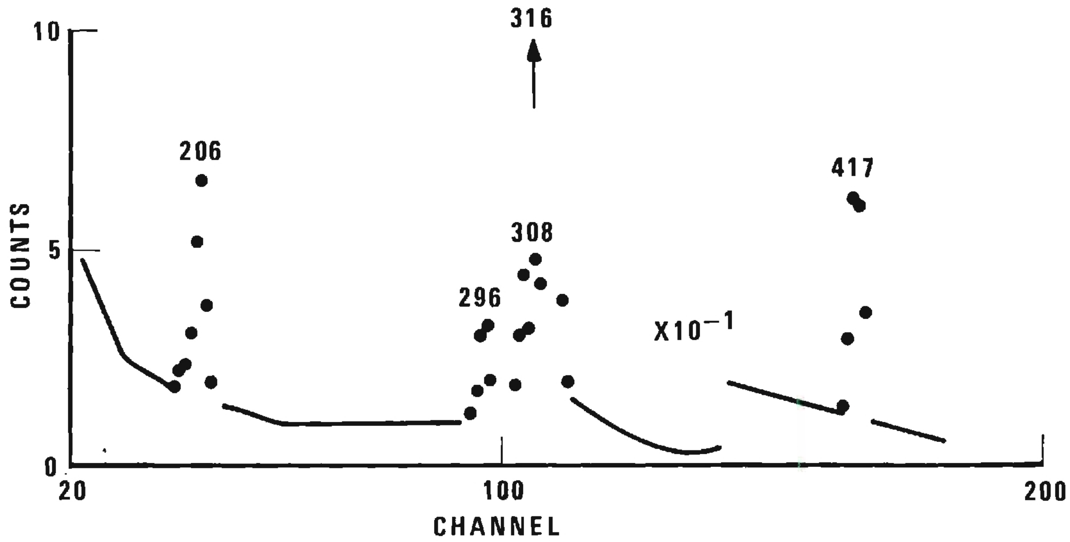

Figure 5a. Coincidence Ge(Li) Spectrum with ~ 470 keV Peak in NaI(Tl) Detector

of real and accidental coincidences is not profitable (20). In general, the counting interval necessary to record a satisfactory coincidence spectrum was two to four hours.

Figure 5b shows the Ge(Li) coincidence spectrum in the ~ 300 keV energy region with the single-channel analyzer set on the 600 keV composite peak. Figure 5a shows the Ge(Li) coincidence spectrum in the energy region (170-500) keV with the single-channel analyzer set on the 470 keV composite peak.

Directional Correlation Measurements

The purpose of a directional correlation measurement is the determination of the correlation coefficients A_2, A_4 --- [equation (1), Chapter I]. Usually it is only necessary to determine A_2 and A_4. [An outline of the gamma-gamma directional correlation theory is discussed in Appendix A.] The following is a brief discussion of problems which occur in gamma-gamma directional correlation experiments (20).

1. Number of angles at which data should be taken. Three angles are sufficient to determine A_2 and A_4. As pointed out in Appendix A, the correlation function, $W(\theta)$, is a function of even order Legendre polynomials; therefore, if data are taken at angles of 225 and 270 degrees, the results should agree with those for 135 and 90 degrees, respectively. Hence a check on certain kinds of experimental asymmetries is obtained by taking data at the five angles 90, 135, 180, 225, and 270 degrees.

2. Source strength. For a given counting time the statistical precision of the data is best if the source strength is such that the

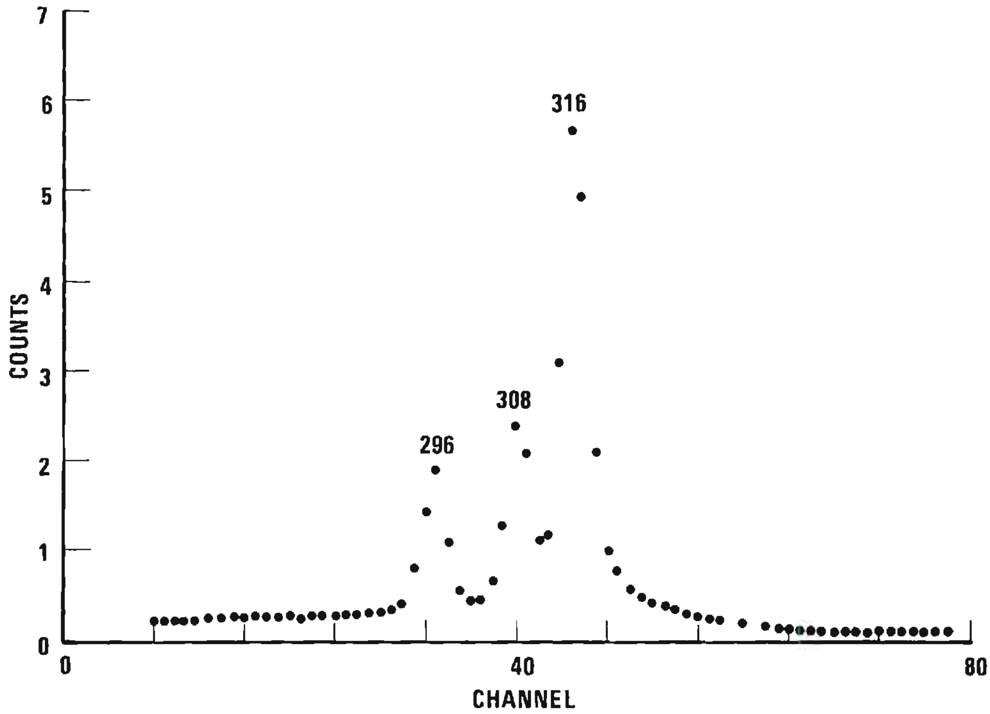

Figure 5b. Coincidence Ge(Li) Spectrum with ~ 600 keV Peak in NaI(Tl) Detector

ratio of the real coincidences to accidental coincidences is approximately unity.

3. <u>Distance from source to detectors</u>. The distance from source to detector determines the solid angle subtended by the detector at the source. The solid angle determines the coincidence rate and the angular resolution of the apparatus. To increase the counting rates the distance must be decreased; to improve angular resolution the distance must be increased. In general, the data may be corrected for the angular resolution of the apparatus; however, the correlation which is observed experimentally before correction tends to be washed out as the angular resolution becomes poorer and, therefore, the detectors cannot be placed too close to the source. A compromise between counting rate and angular resolution must be reached. The distances from the source to the front faces of the Ge(Li) and NaI(Tl) detectors were about 7 cm and 10 cm, respectively.

4. <u>Minimization of systematic errors and effects due to electronic drifts</u>. In order to minimize the systematic errors, data were taken as follows: the coincidence data are taken for a period ranging from two to four hours at one angle and then the NaI(Tl) detectors are moved to another position. Counting was done at five angles per set of runs with the movable NaI(Tl) detectors being positioned relative to the Ge(Li) detector as follows.

D_1	D_2
180	270
135	225
90	180
225	135
180	90
270	180

Note that data were taken twice at 180 degrees in each detector in order to maintain the same statistics at the three independent angles, e.g., 90(270), 135(225), and 180 degrees. The data accumulated at each position of an NaI(Tl) detector are the data for a "run" and the data accumulated for all six runs are called the data for a "set" [of runs].

5. Positioning the source and the Ge(Li) detector. The shape of the Ge(Li) detector is trapezoidal and the problem was whether there was a favored position of the detector at which the singles counting rate could be maximized. Guided by the manufacturer's specifications, Ge(Li) count rates were determined for different relative positions of the source and detector. The position that gave maximum count rate for the 1.28 Mev gamma ray from a test source of Na^{22} was found. The source was centered by ascertaining that the singles count rate of the movable NaI(Tl) detectors was the same at all angles.

6. Analysis of the data. A discussion of the analysis of the data is given in Appendix B. The following adjustments in the coincidence data were taken into account: (i) normalization to the NaI(Tl) singles count rates to correct for the slight shifts in electronics. This also provides some correction for inexact centering of the source and decay of the source (ii) correction for the accidental coincidences due to the finite resolving time of the coincidence circuit (iii) correction due to the coincidence background arising from Compton scattering in the detector (iv) correction due to the finite angular resolution of the apparatus (v) correction due to overlap of two gamma peaks because of the finite resolution of the Ge(Li) detector.

The formula used in data reduction is

$$F_\theta = T_\theta + \frac{T_\theta - 1}{C - 1} + \frac{S'}{S}\left(T_\theta + \frac{T_\theta - 1}{C - 1} - F'_\theta\right)$$

$F_\theta = \frac{N'_\theta}{N'_{90}}$; N'_θ = real coincidence rate at angle θ for the desired cascade

$F'_\theta = \frac{N'_\theta}{N'_{90}}$ for the undesired cascade

$T_\theta = \frac{N_\theta}{N_{90}}$; N_θ = total coincidence rate at angle θ

$C = \frac{N_{90}}{N_{acc}}$; N_{acc} = accidental coincidence rate [average for all angles]

$\frac{S'}{S} = \frac{\text{real coincidence rate due to the undesired cascade}}{\text{real coincidence rate due to the desired cascade}}$.

All count rates, N_θ and N'_θ, are normalized to the singles count rate of the movable NaI(Tl) detector, D_1 or D_2. The correlation coefficients, A_2 and A_4, are determined from $F_{135-225}$ and F_{180}. The geometrical correction factors for the correlation coefficients were approximated for each gamma-gamma cascade. The correction due to the finite energy resolution of the Ge(Li) was applied, where necessary [Appendix B].

The following gamma-gamma directional correlation experiments were performed [gamma energies given in keV]: (468-417), (612-588), (588-296), (612-308), (316-604), (484-206), and (374-206). Table 1 is a summary of the data obtained in each of the above experiments. A discussion of the particulars of each correlation experiment follows.

(468-417) keV correlation experiment. This experiment was performed simultaneously with the (484-206) keV experiment by setting the single-channel analyzers on the composite 470 keV gamma peak and picking out

Table 1. Experimental Gamma-Gamma Directional Correlation Data

NaI D_1 or D_2 + Pb	Cascade NaI	Ge(Li)	N_Π	R	$S_{270/90}$	$S_{225/135}$	T_{180}	$T_{135-225}$	F'_{180}	$F'_{135-225}$	S'/S	A'_2	A_2	A_4
D_1+ 2mm	588	296	20000	2.7	1.00	1.00	0.98 ±0.01	0.99±0.01	0.71	0.86	0.14	-0.02±0.01	0.00 ±0.02	0.00±0.04
D_1+10mm	588	296	17000	3.5	1.02	1.02	0.96 ±0.02	1.00±0.02	0.68	0.84	0.10	-0.03±0.02	0.00 ±0.02	-0.04±0.04
D_2+ 6mm	588	296	25000	3.5	1.05	1.03	0.98 ±0.01	0.99±0.01	0.78	0.93	0.11	-0.02±0.01	-0.01 ±0.02	0.02±0.04
D_1+ 2mm	612	308	23000	3.3	0.98	1.03	0.86 ±0.01	0.95±0.01	0.72	0.88	0.09	-0.13±0.01	-0.12 ±0.02	-0.05±0.04
D_1+10mm	612	308	20000	4.3	0.99	1.00	0.86 ±0.02	0.97±0.02	0.73	0.87	0.06	-0.11±0.02	-0.10 ±0.02	-0.08±0.04
D_2+ 6mm	612	308	35000	4.1	0.96	0.98	0.87 ±0.01	0.97±0.01	0.76	0.92	0.07	-0.11±0.01	-0.10 ±0.02	-0.06±0.04
D_1+ 2mm	308	612	16000	6.1	1.00	1.03	0.86 ±0.03	0.97±0.03	1.10	1.13	0.04	-0.11±0.02	-0.09 ±0.02	-0.07±0.04
D_2+ 2mm	308	612	15000	6.3	1.03	0.97	0.84 ±0.03	0.97±0.03	0.74	0.88	0.03	-0.12±0.02	-0.09 ±0.02	-0.09±0.04
D_1+ 2mm	316	604	18000	7.0	1.02	1.01	0.55 ±0.03	0.80±0.03	1.03	1.11	0.02	-0.45±0.02	-0.48 ±0.02	-0.04±0.05
D_2+ 2mm	316	604	15000	6.0	1.01	0.95	0.52 ±0.03	0.79±0.03	0.77	0.93	0.02	-0.48±0.02	-0.50 ±0.02	-0.06±0.05
D_1+ 2mm	588	612	9500	8.0	1.03	0.97	1.08 ±0.04	1.04±0.04	0.89	0.86	0.11	0.07±0.028	0.082±0.03	-0.02±0.06
D_2+ 2mm	588	612	10000	5.3	1.03	1.03	1.13 ±0.04	1.05±0.04	1.04	0.96	0.06	0.10±0.028	0.12 ±0.03	0.02±0.06
D_1+ 2mm	612	588	9500	6.5	1.02	1.02	1.08 ±0.04	1.02±0.04	1.03	0.92	0.11	0.06±0.028	0.07 ±0.03	0.03±0.06
D_2+ 2mm	612	588	10000	5.9	0.98	0.99	1.09 ±0.04	1.03±0.04	1.01	0.87	0.07	0.07± .028	0.08 ±0.03	0.01±0.06
D_1 NoPb	484	206	21000	6.5	1.03	1.04	0.80 ±0.03	0.91±0.03	1.01	0.99	0.45	-0.18±0.02	-0.27 ±0.03	-0.03±0.06
D_2 NoPb	484	206	30000	6.5	0.95	0.98	0.78 ±0.03	0.93±0.03	1.00	0.99	0.52	-0.18±0.02	-0.28 ±0.03	-0.09±0.06
D_1 NoPb	206	484	18500	57.0	1.01	1.02	0.67 ±0.02	0.88±0.02	1.15	1.09	0.02	-0.26±0.02	-0.27 ±0.02	-0.08±0.04
D_2 NoPb	206	484	25600	50.0	0.98	1.00	0.66 ±0.03	0.86±0.03	1.02	1.01	0.04	-0.26±0.03	-0.29 ±0.03	-0.06±0.05

Table 1. Experimental Gamma-Gamma Directional Correlation Data (Concluded)

NaI D_1 or D_2 + Pb	Cascade NaI	Ge(Li)	N_Π	R	$S_{270/90}$	$S_{225/135}$	T_{180}	$T_{135-225}$	F'_{180}	$F'_{135-225}$	S'/S	A'_2	A_2	A_4
D_1 NoPb	468	417	14000	5.0	1.03	1.04	0.92 ±0.03	0.90±0.03	1.10	1.00	0.11	-0.11±0.02	-0.123±0.03	+0.11±0.06
D_2 NoPb	468	417	21000	5.4	0.99	1.00	0.92 ±0.03	0.90±0.03	1.01	1.07	0.12	-0.11±0.02	-0.118±0.03	+0.11±0.06
D_1 NoPb	206	374	10000	20.0	1.01	0.97	1.120±0.04	1.05±0.04	1.08	1.00	0.35	0.08±0.03	0.10 ±0.03	0.01±0.08
D_2 NoPb	206	374	13800	22.0	1.00	1.01	1.120±0.04	1.05±0.04	1.05	1.01	0.38	0.08±0.03	0.10 ±0.03	0.01±0.08

In the first column, labelled NaI or Ge(Li) + Pb, the amount of lead placed over the faces of the NaI detector, either D_1 or D_2, or the Ge(Li) detector is listed. In the next two columns are listed the energies of the gamma detected by NaI or Ge(Li) counters, respectively. N_Π is the number of coincidence counts for the 180° counting position of the NaI detector. R is the average ratio of the total coincidences to the accidental coincidences. $S_{270/90}$ is the ratio of the anisotropy based on the 270° data to that based on the 90° data. A similar definition holds for $S_{225/135}$. $T_{180} = N_{180}/N_{90-270}$ where, for example, N_{90-270} is the average of the total coincidence count rates at the 90° and 270° counting positions, normalized to the singles count rates of the movable counter. The F'_θ are defined in the discussion of the formula used for data reduction [Appendix B] and the values must be inferred by interpolation since the F'_θ refer to coincidences due to unwanted cascades. S'/S is the ratio of the real coincidences due to the undesired cascades to the real coincidences due to the desired cascade. A'_2 is the second order directional correlation coefficient corrected for accidental coincidences and geometry but not corrected for the coincidence background due to unwanted cascades [i.e., S'/S is equated to zero]. A_2 and A_4 are the final directional correlation coefficients.

the 417 keV peak in the coincidence spectrum with the Ge(Li) detector. Coincident summing of gammas with the NaI(Tl) detectors was not considered important here, so no lead was necessary on the faces of the NaI(Tl) detectors.

The total coincidence counting rate was about 90 counts per hour. The geometrical factors for the correlation coefficients, A_2 and A_4, were taken to be 0.90 and 0.69, respectively.

(612-588) keV correlation experiment. The correlation for this cascade was obtained twice in each measurement, because both 588- and 612-keV were present in the 600 keV gamma group set on the single-channel analyzers and both of these gamma peaks were simultaneously displayed by the Ge(Li) on the 400-channel analyzer. The total coincidence counting rate was about 60 per hour. No correction due to the finite energy resolution of the Ge(Li) was necessary. The geometrical correction factors for the correlation coefficients, A_2 and A_4, were taken to be 0.90 and 0.695, respectively.

(588-296) keV correlation experiment. This experiment was done by setting the single-channel analyzers on the 600 keV composite gamma peak and picking out the 296 keV peak in the coincidence spectrum with the Ge(Li) detector. This correlation experiment was repeated by placing different amounts of lead on the faces of the NaI(Tl) detectors to reduce the possibility of summing the 308- and 316-keV gammas. The total coincidence counting rate ranged from about 350 to 800 counts per hour. The correlation results were found to be about isotropic, as given in Table 1. The 296 keV gamma was completely resolved by the Ge(Li) from the other gammas in the 300 keV group and there was no correction necessary due to the finite resolution of the Ge(Li) detector. The geometrical correction factors for the correlation coefficients, A_2 and A_4, were taken to be 0.893 and 0.681, respectively.

<u>(612-308) keV correlation experiment.</u> This experiment was performed simultaneously with the (588-296) keV correlation experiment. The single-channel analyzers were set on the composite 600 keV gamma peak while the coincidence spectrum of the 308 keV gamma was given by the Ge(Li). The 308 keV gamma was not completely resolved from the intense 316 keV in the Ge(Li) coincidence spectrum. A correction of about four percent was applied to the correlation coefficients by using the calculated correlation coefficients for the composite $\left({}^{588}_{604}\!>\!316\right)$ keV cascade [Appendix B]. Different amounts of lead were placed on the faces of the NaI(Tl) detectors to reduce the possibility of coincident summing of the 296 keV and 316 keV gamma. The total coincidence counting rate was about 750 counts per hour. The geometrical correction factors for the correlation coefficients, A_2 and A_4, were taken to be 0.893 and 0.681, respectively.

The (612-308) keV correlation experiment was also performed with the single-channel analyzers set on the 300 keV composite gamma peak and the 612 keV gamma detected by Ge(Li). The 612 keV gamma was not completely resolved from the 604 keV gamma and about seven percent correction was applied to the correlation coefficients. The total coincidence counting rate was about 350 counts per hour in this experiment. The geometrical correction factors for the correlation coefficients, A_2 and A_4, were taken to be 0.89 and 0.67, respectively. The results of the two correlation experiments on the (612-308) keV cascade agreed to within experimental error.

(316-604) keV correlation experiment. In this experiment, the single-channel analyzers were set on the 300 keV composite gamma peak and the 604 keV gamma was not completely resolved from 612 keV gamma in the Ge(Li) coincidence spectrum and about 2.5 percent correction was applied to the correlation coefficients. The total coincidence counting rate in this experiment ranged from about 300 to 700 counts per hour. The geometrical correction factors for the correlation coefficients, A_2 and A_4, were taken to be 0.89 and 0.67, respectively.

(484-206) keV correlation experiment. The single-channel analyzers were set on the 470 keV composite gamma peak and the coincidence spectrum [Figure 5b] in the energy region of about (170-470) keV was given by the Ge(Li). The contribution from the (489-201) keV cascade is neglected which is a reasonable approximation, because the intensity of this cascade is very much lower than the intensity of the (484-206) keV cascade. This problem is discussed in Chapter V, Suggestions for Future Research. The total coincidence counting rate was about 600 counts per hour. The 206 keV coincidence peak falls on the broad coincidence spectrum due to Compton scattering of the higher energy peaks. Hence the correction due to the coincidence background in this experiment is relatively large. The geometrical correction factors for the correlation coefficients, A_2 and A_4, were taken to be 0.89 and 0.77, respectively.

The (484-206) keV correlation experiment was also performed with the single-channel analyzers set on the 206 keV peak and the 484 keV gamma detected by the Ge(Li). This correlation experiment was done simultaneously with the (374-206) keV experiment. For this experiment, the Ge(Li) was shielded by placing a lead sheet of thickness ~ 4 mm in front of the detector. The lead sheet had a hole of diameter ~ 2 cm so that the Ge(Li) detected only the radiations which came directly from the source. It was found that this additional shielding greatly reduced coincidences due to Compton scattering between counters, especially in the energy region near 374 keV. The total coincidence counting rate was about 200 counts per hour in this experiment. The geometrical correction factors for the correlation coefficients, A_2 and A_4, were taken to be 0.88 and 0.65, respectively. The correction due to the coincidence background in this experiment was very small compared to that applied in the data analysis of the first experiment on the (484-206) keV cascade. The results of the two experiments on the (484-206) keV cascade agreed to within experimental error.

(374-206) keV correlation experiment. The (374-206) keV correlation experiment was performed with the single-channel analyzers set on the 206 keV peak and the 374 keV gamma detected by the Ge(Li). The total coincidence counting rate was about 80 counts per hour in this experiment. The geometrical correction factors for the correlation coefficients, A_2 and A_4, were taken to be 0.88 and 0.64, respectively.

CHAPTER IV

INTERPRETATION OF RESULTS AND CONCLUSIONS

Interpretation of the experimental results is based chiefly on comparison of the experimental correlation coefficients [as given in Table 1] with the theoretical values tabulated by Ferentz and Rosenzweig (2). The final results for the present experiments which have been obtained by a suitable average of the results shown in Table 1 are presented in Table 2. The error limits indicated include the statistical error and some allowance for systematic error in correction for the coincidence background. Additional experimental results, particularly internal conversion coefficients, are considered in cases where appropriate data have been reported in literature. Each gamma-gamma directional correlation experiment, performed in the present research, will be discussed separately with a view to assigning spins for the excited nuclear levels and to fixing limits on the multipolarities of the gamma transitions connecting the various levels [Table 3].

(417-468) keV correlation experiment. Consideration of the decay scheme [Figure 1] shows that the 417 keV gamma, present in the Ge(Li) coincidence spectrum [Figure 5a], is in coincidence only with the 468 keV gamma, present in the 470 keV composite peak set on the single-channel analyzers. This cascade connects the 316-, 785-, and 1200-keV levels in Pt^{192}. The spins of the 316- and 785-keV levels and the multipolarity of the 468 keV transition are well known (20). Therefore, a

Table 2. Final Average Experimental Results

Cascade (keV)	N_{Π}	R	$S_{270/90}$	$S_{225/135}$	A_2	A_4
417-468	35000	5.2	1.01	1.03	-0.12 (3)	0.11 (6)
588-612	39000	6.4	1.02	1.00	0.09 (3)	0.01 (6)
588-296	62000	3.2	1.02	1.02	0.00 (2)	0.00 (4)
308-612	78000	3.9	0.98	1.00	-0.10 (2)	-0.07 (4)
604-316	33000	6.5	1.02	0.98	-0.49 (2)	-0.05 (5)
484-206	95100	60	0.99	1.01	-0.28 (3)	-0.07 (6)
374-206	23800	21.0	1.01	0.99	0.10 (3)	0.02 (6)

Table 3. Summary of Multipolarities of the Gamma Rays Emitted in the Decay of Ir^{192}

E(keV)	Multipolarity	Ref.†	E(keV)	Multipolarity	Ref.†
296	E_2	9	374	E_2	23
	$\delta = -4.4$	10		E_2	*
	E_2	13			
	E_2	19	417	(E_2)	9
	$\delta = -10$	21,26		E_2	23
	$E_2 + M_1$	23		$\lvert\delta\rvert > 10$	*
	$\delta = -6.7 \pm 0.5$	16			
	$\delta = -6.5 \pm 1.5$	12	484	E_2	9
	$\delta = -8.6 \pm 2.0$	31		$\sim 94\% \; E_2 + 6\% \; M_1$	11
	$\delta = 7.0 \pm 2.0$	*		$E_2 + M_1$	23
				$\delta = 11 \pm 2$	31
308	E_2	9		$\delta = 10 \pm 2$	*
	$\delta = -4$	13			
	$\delta = -4$	19	604	$E_2 + M_1$	9
	$\delta = -3$	21,26		$\sim 95\% \; E_2 + 5\% \; M_1$	11
	$E_2 + M_1$	23		$\delta = 3$	13,19
	E_2	12		$\delta = 0.7$	26
	$\delta = -(12 \pm 1)$	31		$\delta = 1.9 \pm 0.4$	31
	$\delta = -7.0 \pm 2.0$	*		$\delta = 2.0 \pm 0.4$	*

†Reference is given according to Bibliography.

*Present investigation.

discussion of the results of the (417-468) keV correlation is relevant to the spin of the 1200 keV level and the multipolarity of the 417 keV transition. A notable feature of the results of this experiment is the large positive value of the A_4 coefficient. If the spin of the 1200 keV level is 3, which was suggested by certain prior information on this decay, the A_4 coefficient should be negative or zero for any mixing of M_1 and E_2 radiations in the 417 keV transition. The present results exclude spin 3 for the 1200 keV level and only spin 4 is a reasonable assignment.

Figure 6a shows a plot of the theoretical directional coefficients A_2 and A_4 versus the mixing parameter f and the experimental value of A_2. The possible values of f are either (i) $f = -0.73 \pm 0.07$; the 417 keV transition is about 53 percent E_2 plus 47 percent M_1 or (ii) 0.995 ± 0.005; the admixture of M_1 radiation in the 417 keV transition is less than two percent. Comparison of the theoretical and experimental values of A_4 indicates that the 417 keV transition contains a strong E_2 component but a choice between the two possible values of f is not possible on the basis of the A_4 coefficient.

Baggerly, et al. (9) and Schellenberg, et al. (23) measured internal conversion coefficients for the 417 keV transition. Comparison of the experimental data with the theoretical values of Sliv, et al. (30) shows that the choice $f = -0.73 \pm 0.07$ must be rejected. The data are consistent with $|f| \approx 1$. The assignment of spin 4 to the 1200 keV level is in agreement with the results of Simons, et al. (14) and Johns, et al. (15), based on gamma-gamma directional correlation measurements.

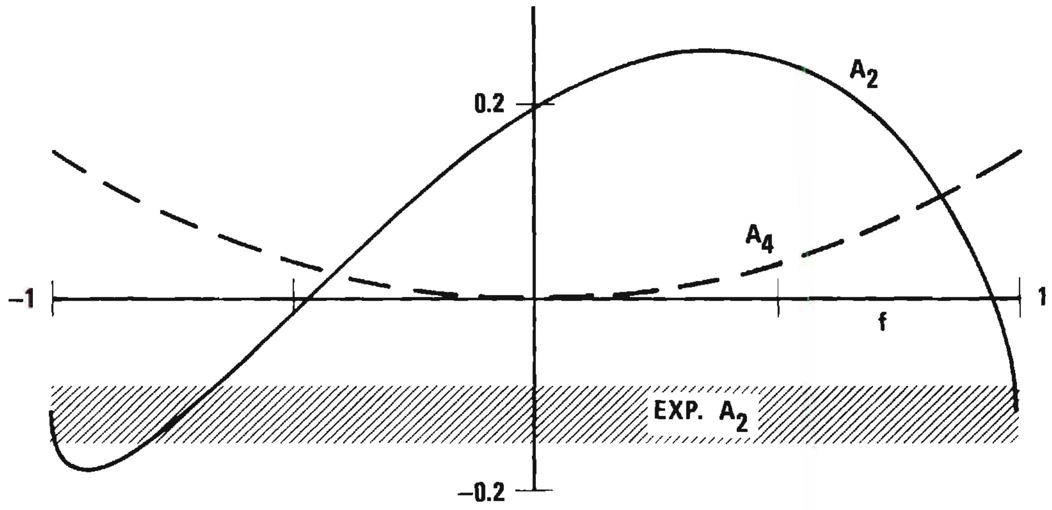

Figure 6a. Plot of the Theoretical Directional Correlation Coefficients vs. the Mixing Parameter for Spin Sequence 4 (E_2 + M_1) 4 (E_2) 2

(588-612) keV coorelation experiment. The present experimental data are in agreement with a 4(2) 2 (2) 0 correlation which should have $A_2 = 0.10$ and $A_4 = 0.01$. These results agree with those of Simons, et al. (14). Shiel, et al. (11) assigned a spin of 3 to the 1200 keV level on the basis of an observed correlation that was approximately isotropic. Kelly, et al. (10) also found the correlation to be approximately isotropic. The data obtained using large NaI(Tl) crystals (14) agree with the present results while the data obtained using small crystals (10,11) evidently show interference effects which tend to destroy the correlation.

(588-296) keV correlation experiment. Consideration of the decay scheme [Figure 1] shows that 296 keV gamma, present in the Ge(Li) coincidence spectrum [Figure 5b], is in coincidence with the 588 keV gamma, present in the 600 keV composite peak set on the single-channel analyzers. This cascade connects the 316-, 612-, and 1200-keV excited levels in Pt^{192}. The spins of the 316- and 612-keV levels are known to be 2. The spin of the 1200 keV level is taken as 4 on the basis of the results obtained from the (417-468) keV correlation. The (588-296) keV correlation is then relevant to the mixing of M_1 and E_2 radiations in the 296 keV transition. Figure 6b shows a plot of the theoretical directional correlation coefficient A_2 versus the mixing parameter f and the experimental value of A_2. The possible values of f are either (i) $f = -0.3 \pm 0.1$; the 296 keV gamma is about 90 percent M_1 plus 10 percent E_2 or (ii) $f = 0.99 \pm 0.01$; the 296 keV gamma is about two percent M_1 plus 98 percent E_2.

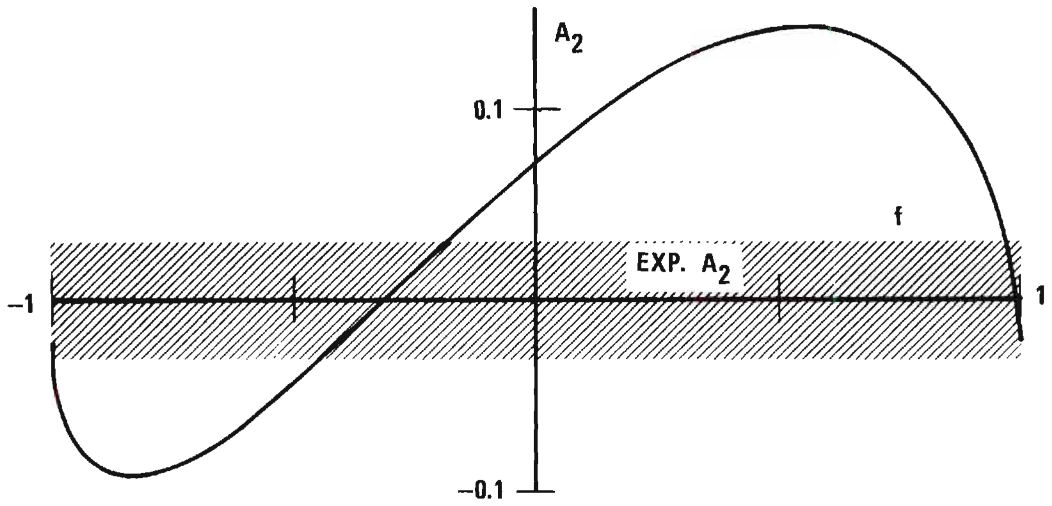

Figure 6b. Plot of the Theoretical Directional Correlation Coefficient vs. the Mixing Parameter for Spin Sequence $4\ (E_2)\ 2\ (E_2 + M_1)\ 2$

K and L sub-shell internal conversion coefficients for the 296 keV transition have been measured by Baggerly, et al. (9), Kelman, et al. (19), and Schellenberg, et al. (23). The theoretical results of Sliv, et al. (30) agree with the experimental conversion coefficients only for the multipolarity of the 296 keV gamma transition to be nearly E_2. Therefore, the multipolarity of the 296 keV gamma as given by $f \simeq -0.3$ is rejected. On the basis of a directional correlation experiment, Grabowski (31) has recently reported $f \approx -0.985$ which would imply an A_2 coefficient somewhat more negative than indicated by the present data.

<u>(308-612) keV correlation experiment</u>. The examination of the data of this experiment is pertinent to a discussion of the spin of the 921 keV level and the multipolarity of the 308 keV transition. The experimental data are not in agreement with a spin of 4 [which, along with spin 3, are the only reasonable spin assignments] for the 921 keV level. For the sequence 4(2) 2 (2) 0, the A_2 coefficient should be 0.102 whereas the experimental value is negative. If the spin is 3, a slight admixture of M_1 radiation in the 308 keV transition is indicated. Figure 6c shows the plot of the theoretical directional correlation coefficient A_2 versus the mixing parameter f. The possible values of f are either (i) $f = 0.04 \pm 0.03$; the 308 keV gamma is almost pure M_1 or (ii) $f = -0.990 \pm 0.005$; the 308 keV gamma is about two percent M_1 plus 98 percent E_2 [the data are not consistent with pure E_2 radiation]. For a dominantly E_2 transition, the A_4 coefficient should be about -0.08, which is consistent with the present experimental result for A_4, while for a dominantly M_1 transition, the A_4 coefficient should be zero, which

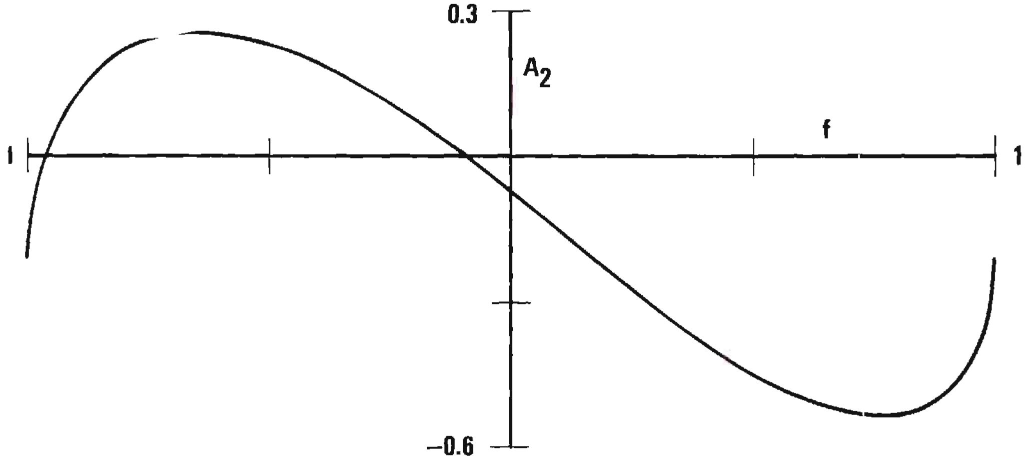

Figure 6c. Plot of the Theoretical Directional Correlation Coefficient vs. the Mixing Parameter for Spin Sequence $3\ (E_2 + M_1)\ 2\ (E_2)\ 0$

is not consistent with the data.

Kelman, et al. (19) and Schellenberg, et al. (23) have studied the conversion electron spectrum with results rather close to those they obtained for the 296 keV transition. Thus, the 308 keV gamma is also dominantly E_2 and the possibility $f \approx 0.04$ is rejected. Grabowski (31) has also recently reported a value for f very close to -1.

(604-316) keV correlation experiment. This experiment provides information on the spin of the 921 keV level and multipolarity of the 604 keV transition. The experimental data preclude a spin of 4 for the 921 keV level [for the sequence 4(2) 2 (2) 0, A_2 = 0.102]. A spin of 3 for the 921 keV level and a substantial admixture of M_1 radiation in the 604 keV transition are in agreement with the present results. Figure 6c shows a plot of the theoretical directional correlation coefficient A_2 versus the mixing parameter f. A conservative conclusion is that f is in the range 0.5 to 0.93.

The conversion coefficient ratios K/L and $L_I:L_{II}:L_{III}$ for the 604 keV transition have been measured by Kelman, et al. (19). Comparison with theoretical values for these ratios (19) indicates a value of f close to 0.9. Grabowski (31) also assigns $f \approx 0.9$ on the basis of a recent directional correlation measurement.

(484-206) keV correlation experiment. This experiment provides information on the spin of the 690 keV level in Os^{192} and the multipolarity of the 484 keV transition. The spins of the ground state and 206 keV level are known to be 0 and 2, respectively, while only spins of 3 or 4 are reasonable values for the 690 keV level. The experimental data preclude the possibility of spin 4 for the 690 keV level [for the sequence

4(2) 2 (2) 0, A_2 = 0.102]. Figure 6c shows a plot of the theoretical directional correlation coefficient A_2 versus the mixing parameter f for the sequence 3(1,2) 2 (2) 0. The possible values of f are either (i) f = 0.26 ± 0.06; the 484 keV gamma is about 93 percent M_1 plus seven percent E_2 or (ii) f = 0.995 ± 0.005; the 484 keV gamma is about two percent M_1 plus 98 percent E_2.

Internal conversion coefficients for the 484 keV transition have been measured by Baggerly, et al. (9) and Schellenberg, et al. (23). Comparison with the theoretical values (30) indicates that the 484 keV transition is predominantly E_2. Hence the choice f ≈ 0.26 is rejected. Shiel, et al. (11) measured the (484-206) keV directional correlation and their experimental data are roughly consistent with the present data. Grabowski's recent report (31) gives f ≈ 1 also.

(374-206) keV correlation experiment. The (374-206) keV cascade connects the 580- and 206-keV excited states and the ground state of Os^{192}. The spins of the ground state and the 206 keV level are known to be 0 and 2, respectively. The experimental data of this experiment are examined in terms of the spin of the 580 keV level and the multipolarity of the 374 keV transition. The present results are in agreement with a spin sequence 4(2) 2 (2) 0 for which A_2 = 0.10 and A_4 = 0.01. Schellenberg, et al. (23) measured the L_{I+II} and L_{III} internal conversion coefficients for the 374 keV transition and found the 374 keV gamma to be E_2, in agreement with the results obtained here.

If the spin of the 580 keV level is taken to be 3, the present experimental data define two possible values of the mixing parameter f for the 374 keV transition [Figure 6c] as either (i) f = -0.19 ± 0.03;

the 374 keV gamma is mainly M_1 radiation or (ii) $f = -0.92 \pm 0.03$; the multipolarity of the 374 keV gamma is about 85 percent E_2 plus 15 percent M_1. Consideration of the results of Schellenberg, et al. excludes the choice $f = -0.19 \pm 0.03$. The directional correlation and the L sub-shell conversion coefficient measurements of Schellenberg, et al. would be consistent with spin 3 and a predominantly E_2 character for the 374 keV transition, in which case the A_4 coefficient should be about -0.08, which is not in accord with the observed value of the A_4 coefficient.

CHAPTER V

SUGGESTIONS FOR FUTURE RESEARCH

The best method of measuring the gamma-gamma directional correlations in Ir^{192} decay would be to use two Ge(Li) gamma detectors, each of 30 cm^3 or more active volume. Each of the gamma peaks would then be cleanly resolved. The movable Ge(Li) detector would have to be especially designed for such an experiment. The suggested experiment might improve the determination of the admixtures of M_1 radiation in the 296-, 308-, 484-, and 604-keV gamma transitions.

The (206-283) keV correlation experiment, attempted in the present work, involves a number of problems. The 283 keV transition is very weak compared to the intense peaks of the 300 keV group (25). The weak 206 keV gamma peak, though resolved in the singles spectrum of a 3 x 3 inch NaI(Tl) detector, falls on the Compton background formed by the higher energy peaks. At present, one possible way the (206-283) keV correlation experiment can be performed is by setting a 3 x 3 inch NaI(Tl) detector on the 206 keV gamma peak and detecting the 283 keV gamma by the Ge(Li) detector. However, coincidences are produced by ~ 300 keV gammas in the Ge(Li) detector, which are not cleanly resolved from the 283 keV gamma and various higher energy gammas which undergo Compton scattering in the NaI(Tl) detector. Correction for this coincidence background is difficult. Two Ge(Li) detectors can be tried in this experiment, but the lower efficiency of the Ge(Li) detector will cause the rate of data acquisition to be very slow.

A second possibility, which seems to be more feasible, is to set the (283-316) keV composite peak on the NaI(Tl) detectors. The coincident 206 keV gamma peak is then detected by the Ge(Li). In experiments on cascades involving one or both gammas of energy lower than the other intense gammas, there is a serious difficulty due to Compton scattering between counters (29). It may be necessary to shield the Ge(Li) detector from the other detectors.

For rapid data acquisition, especially in the case of cascades of weak gamma peaks, it will be desirable to use a multichannel analyzer of about a thousand channels so that the entire spectrum in coincidence with a particular gamma is recorded in one experiment. An automatic system of data recording and angle changing is also recommended to allow continuous operation of the experiment.

During this research, the presence of the 690-, 785-, and 921-keV gammas was detected in the Ge(Li) singles spectrum of Ir^{192}. The existence of these gammas cannot possibly be confirmed by gamma-gamma directional correlation experiments. These peaks could result due to sum peaks in the detector. It is suggested that a study of the decay scheme of Ir^{192} be undertaken employing a 30 cm^3 or bigger Ge(Li) detector with a suitable Ge(Li)-source geometry.

APPENDICES

APPENDIX A

THEORY OF GAMMA-GAMMA DIRECTIONAL CORRELATIONS

This appendix is an outline of theoretical results appearing in the literature and relevant to the present research. For a comprehensive treatment of the theory, the reader is referred to the article by H. Frauenfelder and R. M. Steffen (2). Before this theoretical outline is given, it is worthwhile to explain some basic properties of nuclear states and nuclear radiation.

Basic Properties of Nuclear States and Nuclear Radiation

A nucleus is a bound system of nucleons [neutrons and protons]. This system seems to obey the laws of quantum mechanics and so it should have a set of quantized energy states, the ground state, and the excited states. A nuclear state is characterized by energy, total angular momentum or spin, and parity (20). The energies of the excited nuclear states are determined by measuring the energies of the gamma rays connecting the states. Gamma radiations are conveniently classified by multipole order L, according to the angular momentum [in units of $\hbar$] carried by each quantum of radiation (32). The possible classifications are dipole, quadrupole, octopole, . . . radiations corresponding to values of L being 1, 2, 3, . . ., respectively. Each type of gamma radiation is further classified as 2^L-pole electric (E_L) or 2^L-pole magnetic (M_L) depending on parity (Π).*

*Parity (Π) refers to the symmetry property of the system wave function under inversion of the coordinates.

The laws of conservation of angular momentum and of parity of the system [nucleus plus gamma] restrict the multipolarity of a gamma transition. In a gamma transition connecting initial and final states of spin quantum numbers J_i and J_f, only certain types of transitions are allowed by the following selection rules.

$$|J_f - J_i| \leqq L \leqq J_f + J_i \qquad \text{and}$$

$$\Delta\pi \equiv \pi_i/\pi_f = (-1)^L \qquad \text{for } E_L \text{ radiation}$$

$$= (-1)^{L+1} \qquad \text{for } M_L \text{ radiation}$$

where

$\Delta\pi = +1$ represents no parity change

$\Delta\pi = -1$ represents a parity change

In general, the transition probability for a gamma transition decreases very rapidly with increasing multipolarity L and for most practical purposes, all but the two lowest possible L-values are neglected. The law of conservation of parity excludes the possibility of simultaneous electric and magnetic gamma transitions of the same multipolarity.

Gamma-Gamma Directional Correlations

The directional correlation function $W(\theta)$ is developed here. Let a nucleus decay through successive emission of two electromagnetic quanta [gamma rays] in directions $\vec{k_1}$ and $\vec{k_2}$. Let these gamma rays be emitted during two successive transitions of the nucleus connecting three nuclear states with spins J_1, J_2, and J_3. Each nuclear state, represented by $|J\rangle$, is degenerate with regard to the magnetic quantum number m and is associated with $(2J + 1)$ substates represented by

$|Jm>$. The directional correlation function $W(\theta) = W(\vec{k_1} \cdot \vec{k_2})$ is defined as the relative probability that two nuclear radiations will be emitted with angle θ between their propagation directions.

Hamilton (4) developed an expression for $W(\vec{k_1} \cdot \vec{k_2})$ using second order time dependent perturbation theory for an initial system of excited nucleus and quantized radiation field. Hamilton calculated the probability that a given nucleus will be found in its final state f after decaying and that two gamma rays will have been emitted with definite directions, energies, and polarizations. The function $W(\vec{k_1} \cdot \vec{k_2})$ follows from this probability after averaging over all nuclei, all gamma ray energies, all final nuclear states, and all unresolved properties of the quanta (2). The general form of $W(\theta)$ is given by

$$W(\theta) = S_1 S_2 \sum_{m_1 m_3} \left[\sum_{m_2} (J_1 m_1 | H(\vec{k_1}) | J_2 m_2)(J_2 m_2 | H(\vec{k_2}) | J_3 m_3) \right]^2 \qquad (1)$$

$H(\vec{k_1})$ and $H(\vec{k_2})$ are the interaction Hamiltonians of the emission of the first and second gamma rays in the directions $\vec{k_1}$ and $\vec{k_2}$, respectively. S_1 and S_2 indicate summation over all unmeasured properties of gamma rays like spin and polarization. Only the direction of propagation of the particles is observed. The matrix elements are the probability amplitudes for the various possible transitions between m-degenerate states.

Let the quantization axis be chosen along $\vec{k_1}$, the direction of propagation of the first radiation, then

$$W(\theta) = \sum_{m_2} \left\{ \sum_{m_1} \left[S_1 \left| (J_1 m_1 | H(0) | J_2 m_2) \right|^2 \right] \right. \tag{2}$$

$$\left. \times \sum_{m_3} \left[S_2 \left| (J_2 m_2 | H(\theta) | J_3 m_3) \right|^2 \right] \right\} \equiv \sum_{m_1 m_2 m_3} P_{m_1 m_2}(0)\ P_{m_2 m_3}(\theta).$$

$P_{m_1 m_2}(0) = S_1 \left| (J_1 m_1 | H(0)_1 J_2 m_2) \right|^2$ gives the relative probability that the first radiation is emitted along the direction $\theta = 0$ during the nuclear transition $m_1 \rightarrow m_2$. A similar definition applies to $P_{m_2 m_3}(\theta)$.

Angular Distribution Functions

The Hamiltonian $H(\theta)$ in $P_{m_2 m_3}$ is invariant under any rotation of space coordinates and must have the form (33)

$$H = \sum_{M=-L}^{L} H_{L,M}\ (\vec{\nu}_i, \vec{x}_i) \tag{3}$$

$$= \sum_{M} (-1)^M A_{L,-M}\ (\vec{\nu}_i)\ T_{L,M}\ (\vec{x}_i)$$

where M is the component of L along quantization axis. The tensor $T_{L,M}$, of rank L, depends on the coordinates, $\vec{x}_i$ [operators], of the nucleus and $A_{L,-M}$, of rank L, depends on the variables, $\vec{\nu}_i$, that describe the radiation field. For gamma radiation, $T_{L,M}$ consists of multipole moments obtained from the charge, current, and magnetization densities in the nucleus and the $A_{L,M}$ are derived from the vector spherical harmonic expansion of the electromagnetic field (33).

Putting the expression for H into $P_{m_2 m_3}$, the nuclear matrix elements become

$$(J_2 m_2 | H_{L,M} | J_3 m_3) = (-1)^M A_{L,-M} (J_2 m_2 | T_{L,M} | J_3 m_3). \quad (4)$$

The factor $A_{L,-M}$ does not depend on the nuclear coordinates so it can be removed from the nuclear matrix element. Using the Wigner-Ekart theorem (1) the nuclear matrix element in equation (4) is given by

$$(J_2 m_2 | T_{L,M} | J_3 m_3) = (J_2 m_2 LM | J_3 m_3)(J_2 || T_L || J_3) \quad (5)$$

where $(J_2 || T_L || J_3)$, the reduced matrix element of the tensor operator $T_{L,M}$, is independent of m_2, m_3, and M. The conservation of angular momentum is contained in the vector addition coefficient $(J_2 m_2 LM | J_3 m_3)$ [Clebsch-Gordan coefficient] which vanishes unless $m_3 = m_2 + M$. Hence the sum in equation (3) is a single term.

We then have

$$P_{m_2 m_3}(\theta) = S_2 |(J_2 m_2 | H(\theta) | J_3 m_3)|^2$$

and [ignoring the reduced matrix element which is common in each component $m_2 \rightarrow m_3$ of the transition $J_2 \rightarrow J_3$] the probability is given by

$$P_{m_2 m_3} \propto (J_2 m_2 LM | J_3 m_3)^2 \, F_{L,M}(\theta)$$

where

$$F_{L,M}(\theta) = S_2 |A_{L,M}(\vec{v}_i)|^2$$

which gives the angular distribution function of the gamma radiation corresponding to quantum numbers L and M. S_2 represents the sum over all vectors $\vec{v}_i$ that describe the radiation except $\vec{k}_2$, the direction of emission of the particle. For spinless particles like alpha particles, only the propagation vector $\vec{k}_2$ is needed to describe the particle and the summation S_2 is not required.

The general form of the tensors $A_{L,M}$ is

$$A_{L,M}\ (\vec{\nu}_i) = a_L\ (\nu_i)\ y_{L,M}\ (\theta,\Phi)$$

Hence for spinless particles

$$F_{L,M}\ (\theta) \propto \left|y_{L,M}\ (\theta,\Phi)\right|^2 .$$

The azimuthal dependence disappears in the squaring of the spherical harmonics.

For gamma radiation, a sum over the polarization vectors is required. $F_{L,M}\ (\theta)$ can be calculated by evaluating the Poynting vector as a function of θ for the multipole radiation characterized by L and M (32) and is given by

$$F_{L,M}(\theta) = \frac{1}{2L(L+1)} [\,(L+M)(L-M+1)\left|y_{L,M-1}(\theta,\Phi)\right|^2 + M^2\left|y_{L,M}(\theta,\Phi)\right|^2$$

$$+ (L-M)(L+M+1)\left|y_{L,M+1}(\theta,\Phi)\right|^2\,] .$$

The Directional Correlation Function $W(\theta)$

Exhibition of the explicit angular dependence of $W(\theta)$ is aided by employment of the techniques of Racah algebra as presented by Frauenfelder and Steffen (2). A discussion of the more general theory is not presented here but some of the results will be mentioned.

The correlation function $W(\theta)$ for gamma-gamma directional correlations is given by a sum of a finite number of even order Legendre polynomials

$$W(\theta) = \sum_{n\ \mathrm{even}} A_n P_n (\cos\theta)$$

where

$$n_{max} = \min (2J_2,\ 2L_1,\ 2L_2)$$

L_1 and L_2 are the angular momenta of the first and second radiations and J_2 is the spin of the intermediate nuclear state. A_ν is further broken into two factors each depending upon only one transition of the cascade

$$A_\nu = F_\nu^{(1)} (J_2 J_1 L_1 L_1)\ F_\nu^{(2)} (J_3 J_2 L_2 L_2).$$

These factors depend only on the spins and multipolarities of the first and second transitions of the cascade. These factors have been numerically tabulated by Biedenharn and Rose (7) and by Ferentz and Rosenzweig (2).

For the case where either or both gamma transitions in the cascade are of mixed multipolarity, the theory has been extended and $F_\nu^{(1)}$ is given by

$$F_\nu^{(1)} = (1-f_1^2)\ F_\nu\ (J_2 J_1 L_1 L_1) + 2f_1\sqrt{1-f_1^2}\ F_\nu\ (J_2 J_1 L_1 L_1')$$

$$+ f_1^2\ F_\nu\ (J_2 J_1 L_1' L_1')$$

where

$$f_1^2 = \frac{\delta_1^2}{1 + \delta_1^2}$$

$$L_1' = L_1 + 1$$

and a similar expression holds for $F_\nu^{(2)}$ (2). The [amplitude] mixing ratio, δ_1, of the transition 1 is defined as the ratio of the reduced matrix elements

$$\delta_1 \equiv < J_2 \| L_1' \; \Pi_1' \| J_1 > / < J_2 \| L_1 \; \Pi_1 \| J_1 >.$$

The ratio of the intensity of the L'-pole radiation to that of the L-pole is equal to δ_1^2. The reduced matrix elements for the gamma transition can always be chosen to be real; therefore, the mixing ratio δ_1 is real. For a given intensity ratio δ_1^2, the mixing ratio δ_1 can have either a positive or a negative sign depending on the relative phase of the reduced matrix elements. By definition

$$f_1 = \frac{\delta_1}{\sqrt{1 + \delta_1^2}}, \quad -1 \leqq f_1 \leqq 1.$$

The value of f_1^2 gives the relative contribution of the radiation of multipolarity L_1' in the transition 1. A similar discussion holds for the mixing ratio δ_2 of the transition 2.

APPENDIX B

ANALYSIS OF EXPERIMENTAL DATA AND CORRECTIONS

The experimental data were analyzed to determine the coefficients in the correlation function [defined in Chapter I]

$$W(\theta) = 1 + A_2 P_2 (\cos\theta) + A_4 P_4 (\cos\theta).$$

Table 4 contains a sample of the experimental data and analysis for directional correlations in the (612-308) keV cascade. The following procedure for data analysis was used.

1. The average singles counting rate [per hour], S, of the NaI(Tl) detector D is calculated for each run [angular position θ].

2. The average total [real plus accidental] or accidental coincidence count rate of the Ge(Li) - NaI(Tl) system is calculated for each run by summing the total coincidences over the three or four channels in the 400-channel analyzer that span the desired peak. This coincidence rate is denoted by N.

3. The ratio N/S is calculated for each run.

4. The average of the quantity N/S is calculated for each angular position (θ) and is denoted by $(N/S)_{90-270}$; $(N/S)_{135-225}$; $(N/S)_{180}$.

5. T_{135} and T_{180} are obtained for each set of the data as follows

$$T_{135} = \frac{(N/S)_{135-225}}{(N/S)_{90-270}}, \quad T_{180} = \frac{(N/S)_{180}}{(N/S)_{90-270}}.$$

Table 4. Sample of Data and Analysis for Directional Correlations in [~ 600 ⋞ 300] keV Cascades

The ~ 600 keV composite peak is set on the NaI(Tl) [D_2] detector and the 308 keV gamma is detected by the Ge(Li) detector. It is necessary to analyze the $\left(\begin{smallmatrix}588\\604\end{smallmatrix} > 316\right)$ keV composite cascade for the correction due to the overlap of the 308- and 316-keV gamma peaks because of the finite energy resolution of the Ge(Li). A_2' and A_4' are uncorrected for the finite energy resolution; A_2 and A_4 are the final results.

θ	$G = \frac{\text{Total Coincidence Rate}}{\text{NaI(Tl) Singles Rate}}$ (308 keV)	(316 keV)
90°	19.617	41.010
135°	18.565	35.752
180°	17.338	27.359
225°	18.459	34.387
270°	19.297	42.536
180°	16.931	27.222

Peak (keV)	G 90-270°	G 135-225°	G 180°	T_{135}	T_{180}	C	$F_\theta^1 = T_\theta + \frac{T_\theta - 1}{C - 1}$ (θ=135°)	(θ=180°)	S'/S	$F_\theta'(E_0)$ (θ=135°)	(θ=180°)
308	19.457	18.512	17.135	0.951	0.881	4.29	0.94±0.01	0.84±0.01	0.07	0.92	0.76
316	41.773	35.070	27.291	0.840	0.653	4.99	0.80±0.01	0.57±0.01	0.02	0.92	0.76

$F_\theta = F_\theta^1 + S'/S\cdot(F_\theta^1 - F_\theta'(E_0))$ (θ=135°)	(θ=180°)	A_2'	A_4'	A_2	A_4
0.94±0.01	0.85±0.01	-0.11±0.01	-0.02±0.02	-0.10±0.01	-0.02±0.02
0.80±0.01	0.56±0.01	-0.37±0.01	-0.03±0.02		

6. After the quantities T_{135} and T_{180} are calculated for all the sets of data, the average of these quantities is obtained and the statistical errors are estimated.

7. The value of C is obtained as follows

$$C = \frac{\text{Average } (N/S)_{90-270} \text{ for total coincidences}}{\text{Average } (N/S) \text{ for accidentals [at all angles]}}.$$

8. For a chosen set, a plot of average total coincidence rate at 90° and 270° against channel number is drawn. The total coincidence count rate, n, over the prescribed number of peak channels due to the coincidence background is estimated by interpolation.

9. A plot of average accidental count rate over all angles against channel number is drawn. The total accidental count rate, a, over the prescribed number of peak channels due to the accidental background is estimated by interpolation.

10. S' is obtained as follows: $S' = n - a$.

11. The average total coincidence count rate, N, at 90° and 270° for the same set and over the same prescribed number of peak channels is obtained.

12. The average accidental count rate, A, at all angles and over the same prescribed number of peak channels is obtained.

13. S is obtained as follows: $S = N - A - S'$.

14. S'/S is calculated.

15. $F'_\theta(E_0)$ is obtained by interpolation from plots of F'_θ against channel number. F'_θ is calculated for energies [channel numbers] somewhat lower and higher than the energy E_0, of the desired gamma peak

detected by the Ge(Li) according to

$$F'_\theta = T_\theta + \frac{T_\theta - 1}{C - 1}.$$

16. F_{135} and F_{180} are calculated from the formula used in the data reduction

$$F_\theta = T_\theta + \frac{T_\theta - 1}{C - 1} + \frac{S'}{S}\left(T_\theta + \frac{T_\theta - 1}{C - 1} - F'_\theta(E_0)\right).$$

17. Using F_{135} and F_{180}, the correlation coefficients are determined as follows

$$A_2 = \frac{0.78125\ F_{180} + 0.625\ F_{135} - 1.40625}{0.109375\ F_{180} + 0.875\ F_{135} + 0.65625}$$

$$A_4 = \frac{0.75\ F_{180} - 1.5\ F_{135} + 0.75}{0.109375\ F_{180} + 0.875\ F_{135} + 0.65625}.$$

These values of A_2 and A_4 are further corrected for the finite angular resolution of the gamma detectors. A method of estimating these geometrical corrections has been discussed by Rose (34) and Yates (2). The required corrections are made by dividing the correlation coefficients A_2 and A_4 by certain correction factors Q_2 and Q_4, respectively. Each Q_i is a product of two factors corresponding to the NaI(Tl) and the Ge(Li) detectors, respectively: $Q_i = Q_i(1)\ Q_i(2)$. These correction factors depend on the source-detector distance h, diameter 2r, thickness t of the detector and energy E_0 of the gamma radiation. Values of Q_2 and Q_4 for 3 x 3 inch NaI(Tl) detectors are tabulated in reference (2).

The values of Q_2 and Q_4 for the Ge(Li) detector were estimated by measuring the correlation coefficients A_2 and A_4, uncorrected for finite angular resolution of the detectors, for the (570-1060) keV cascade in Bi^{207}. It was found that the correction factors are about the same as those for a 1.5 X 1 inch NaI(Tl) detector. This approximate equivalence of the correction factors is reasonable in view of the geometry of the Ge(Li) detector. More precise correction factors for the Ge(Li) detector are not available at present but it is not thought that the error due to this cause is serious. A further check on the corrections follows from the fact that the correlation experiments on the (308-612) keV cascade were done twice; first, the 612 keV gamma was detected by the NaI(Tl) detectors and then by the Ge(Li) detector. The correlation results agreed to within the experimental error.

The values of Q_2 and Q_4 used for the various experiments are quoted in Chapter III.

In correlation experiments on the (316-604) keV and (308-612) keV cascades, a correction due to the finite energy resolution of the Ge(Li) detector was necessary [see Table 4]. The estimate of the correction was made by constructing curves which represented the observed spectrum in the vicinity of the 600- and 300-keV gamma peaks. This was done by assuming that each gamma ray yields a Gaussian distribution, $G(E_0)\ e^{-(E-E_0)^2/k}$, characterized by a peak height, $G(E_0)$, proportional to gamma intensity (25) and width, ΔE, at half maximum dependent upon the resolution of the Ge(Li) at the gamma energy E_0

$$k = (\Delta E)^2/2.772$$

$$\Delta E = E_0 \times R/100$$

$R \equiv$ resolution in percentage of the Ge(Li); found for the 316 keV gamma.

The Gaussian curves for individual gamma peaks were drawn. The curves for the composite gamma peaks, 300- and 600-keV, were drawn by algebraic summing of the individual peaks. The estimate [in percent] of contribution of a neighboring peak to the other in the coincidence spectrum is made from the degree of overlap of these peaks. For example, in the correlation experiment (612-308) keV, in which the 600 keV composite gamma is detected by the NaI(Tl) detectors and 296-, 308-, and 316-keV gammas by the Ge(Li), the correction is made for contribution of the 316 keV gamma to the 308 keV gamma in the coincidence spectrum as follows. $(A_i)_{uncorrected}$ is found for the (612-308) keV cascade and A_i is found for the composite cascade $\left(316 < {588 \atop 604}\right)$ keV. The contribution, x percent, of 316 keV peak to the 308 keV peak is determined, as explained above. The $(A_i)_{corrected}$ for the (612-308) keV cascade is obtained from

$$(A_i)_{corrected} = [(A_i)_{uncorrected} \text{ for (612-308) keV cascade}$$

$$-(x/100)\, A_i \text{ for } \left(316 < {588 \atop 604}\right) \text{ keV cascade} \,/\, [1-(x/100)]].$$

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VITA

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